Analysis of organic contaminants in New Zealand marine mammals

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Abstract

1. Chlorinated organic contaminants were analysed in samples of blubber collected from New Zealand's Hector's dolphin (*Cephalorhynchus hectori*) and dusky dolphins (*Lagenorhynchus obscurus*).

2. Polychlorinated biphenyl (PCB) congeners were detected in all samples analysed. Total PCB concentrations measured in these samples were lower than those detected in cetaceans from other countries.

3. Polychlorinated dibenzo-*p*-dioxin (PCDD) and dibenzofuran (PCDF) congeners were also detected in many of the samples. These compounds were more common and present at higher concentrations in Hector's dolphins than in the dusky dolphin.

4. Data from this study will be used to perform pattern recognition analysis of the contaminants detected. Using these methods we hope to be able to better identify source areas for these contaminants.

5. It is difficult to predict adverse effects of contaminants on the dolphins at the level of the individual or population level. The fact that contaminant concentrations detected in New Zealand dolphins are considerably lower than concentrations in northern hemisphere cetaceans suggests some margin of safety.

1. Introduction

Limited information is available on the occurrence and distribution of planar chlorinated hydrocarbons (PCHs) such as polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in the southern hemisphere. PCHs are known to bioaccumulate and biomagnify, and recent studies have linked these compounds to reproductive deficiencies in some wildlife species. Marine mammal species have been shown to bioaccumulate PCHs to high concentrations [1,2] and are therefore at particular risk from the effects of these contaminants. This ability to accumulate high levels of PCHs also makes marine mammals excellent sentinel species for contamination of the environment by PCHs [2].

Recent studies have indicated that marine mammals around the New Zealand coast are exposed to and accumulate considerable concentrations of planar chlorinated hydrocarbons. Current work is focusing on the use of pattern recognition techniques to evaluate possible sources of these contaminants. To date this approach has been successful in identifying a contaminant profile indicative of exposure in the open ocean environment. This profile is characterised by very low levels of PCDD and PCDF and an increased prevalence of lower chlorinated PCB congeners. In contrast, the contaminant pro-

file from inshore feeders shows a 'normal' abundance of lower chlorinated PCBs and an increase in the levels of PCDD and PCDF congeners relative to PCBs[3].

We are now using these pattern recognition techniques to attempt to identify different contaminant sources around the New Zealand coast. This approach has been used in US studies. However, interpretation of these studies is complicated by the relatively large home ranges of the marine mammals studies. In this respect Hector's dolphin (*Cephalorhynchus hectori*) is a particularly interesting species to study because it has a very small home range. In addition we are able to compare contaminant profiles from Hector's dolphin with open-water species such as the common dolphin (*Delphinus delphi*) and dusky dolphin (*Lagenorhynchus obscurus*).

In this study we have analysed samples of dolphin blubber from around the New Zealand coast. A total of 11 dolphins (8 Hector's dolphins and 3 dusky dolphins) have been analysed for PCDD and PCDF congeners. The same 11 dolphins along with another 4 Hector's dolphins have also been analysed for PCB congeners.

2. Methods

Samples were obtained from various sources affiliated with the Department of Conservation. Samples were stored frozen at -20 °C until analysis. Biometric data for the individual dolphins is provided in Appendix 1.

PCDD, PCDF and PCB congeners were analysed as described previously[3,4]. Samples were fortified with $^{13}C_{12}$ PCDD, PCDF and PCB congeners and were extracted four times by blending with 30 ml 2:1 acetone:hexane. Extracts were dried by passage though anhydrous Na_2SO_4, concentrated to near dryness and redissolved in 50 ml of hexane. A 5.0 ml portion of the extract was removed for gravimetric lipid determination. The remaining extract,after any sub-sampling, was transferred to a separating funnel and washed repeatedly with concentrated H_2SO_4 (8 times) followed by repeated washing with H_2O (3 times). The extract was again dried through Na_2SO_4 before being chromatographed sequentially on columns of H_2SO_4 silica: NaOH/silica, Al_2O_3, and Carbopac C dispersed on Celite. PCB congeners pass through the Carbopac column while PCDDs and PCDFs are retained and eluted into a separate fraction. The PCB fraction was chromatographed on Florisil to isolate the three non-ortho substituted congeners. All analytes were determined by HRGC/HRMS on a VG70S mass spectrometer.

Full analytical details are available on request

3. Quality assurance

The dioxin laboratory maintains WHO and TELARC accreditation for the analysis of PCDD, PCDF and PCB congeners in a variety of environmental matrices. Laboratory blanks were run with each batch of samples. All data analysis was subject to strict quality assurance procedures.

4. Results

PCB congeners were detected in all samples analysed (Appendix 2). The sum of the 19 congeners analysed varied from 0.37 to 4.17 μg/g Previous comparative data suggests that the congeners analysed represent, on average, 32% of the total PCB concentration. Therefore total PCB concentrations in these samples can be estimated to range from 1.2 to 14.4 μg/g.

The three most biologically potent PCBs are PCB#77 PCB#12 and PCB#169, all of which owe their potency to their ability to assume a planar molecular configuration[5]. The three coplanar PCB congeners were detected in all samples analysed. These congeners are less abundant than the other PCBs (their concentrations are reported in pg/g). However, due to their biological potency, these and two other more abundant but less potent congeners, PCB#105 and PCB#118, account for most of the biological potency of the PCB mixtures.

PCDD and PCDF congeners were also detected in many of the samples analysed (Table 3). Actual 2, 3, 7, 8-TCDD was detected at a highest concentration of 7.02 pg/g. PCDD and PCDF concentrations were lower in the open-ocean dusky dolphins than in the inshore Hector's dolphin. This phenomenon has been demonstrated for other species of open-ocean cetacean, [3] and probably demonstrates the limited distribution of these compounds in the atmosphere as compared to PCBs.

Since PCDDs, PCDFs and PCBs are believed to act through the same receptor, mediated mechanism, it is possible to calculate the total dioxin like potency of a mixture of these contaminants by using toxic equivalency factors (TEFs) for each compound[5]. This approach has been proven to be useful in estimating the potential for environmental mixtures of contaminants to cause adverse effects. This method also allows a comparison of the relative risk of the different groups of compounds.

For the calculation of toxic equivalents (TEs), the concentrations of individual congeners in the sample are multiplied by their TEF value, which measures their biological potency relative to dioxin. The TE contributed by each congener in a sample can then be added to give a total TE concentration for that sample. There is currently considerable effort going into determining the appropriate TEF values for the different congeners, since there appears to be significant variation in TEFs between species and between different biologi-

cal endpoints[5]. To overcome these difficulties we chose to use the TEF values currently used for human health risk assessment [6,7].

Total TE concentrations in the samples analysed varied from 0.3 to 200 pg/g (Appendix 3) and total TE concentrations were lower in the dusky dolphins than in Hector's dolphin. In addition TEs in the dusky dolphins were almost exclusively derived from PCB congeners (Appendix 3).

5. Discussion

The concentrations of PCBs found in the New Zealand marine mammals analysed were considerably lower than levels detected overseas, particularly in the northern hemisphere [8] **(Appendix 4).** In addition, concentrations detected in the dusky dolphins were similar to those previously reported for this species[8]. Concentrations detected in Hector's dolphin were similar to those previously analysed[9].

Few data are available on PCDD and PCDF in marine mammals. High concentrations of PCDD, PCDF and PCB congeners have been reported in killer whale[10] while considerably lower concentrations have been reported in Hector's dolphins. Levels reported here are similar to those previously reported from Hector's dolphin[4].

The major aim of this project was to provide additional data for the pattern analysis of contaminant profiles in Hector's dolphin. The addition of another 11 dolphins to the data currently held by ESR will greatly facilitate this analysis. It is obvious from the data presented here that there is a marked difference in the contaminants found in Hector's dolphin compared with open-ocean species. Using these data we hope to be able to identify contaminant profiles characteristic of Hector's dolphins from specific locations. If this analysis is successful we will be well on the way to identifying possible contaminant sources. This complex statistical analysis using these data is under way.

The major question arising from these findings is what effect these contaminants could be having on the dolphins. Unfortunately this question is very difficult to answer. Species vary greatly in their sensitivity to PCHS; even closely related species can vary more than 1000 fold in their sensitivity to these compounds. For this reason it is necessary to compare known effect levels in a variety of similar species to attempt to infer effect levels in the species of interest. As many of the necessary effect data need to be generated in controlled exposure experiments, this type of data is practically non-existent for cetacean species. There is evidence that exposure to PCHs causes immunological and reproductive anomalies in cetaceans [11], but much of this evidence is circumstantial and does not provide solid scientific evidence of a cause/effect relationship[12,13].

We are aware of only one study that has demonstrated a measured biological effect of exposure to PCHs and even in this case the effect was not statistically significant[14]. In this study a trend toward decreased blood sex hormone levels was observed in male Dall's porpoise with blubber concentrations of PCBs ranging from 5.6 to 17.8 μg/g. These concentrations are within the range of those detailed in this report suggesting that New Zealand dolphins may be experiencing some effects of these contaminant burdens. It cannot be determined if these effects will have an impact at the level of the whole animal or population.

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Appendix 1. Biometric data for the individual mammals analysed.

Lab#	Doc#	Species	Sex	Age (y)	Location
MM8-HD2	9162	Hector's	Female	1	Waikouaiti
MM8-DD1		Dusky			Kaikoura
MM8-DD2		Dusky			Kaikoura
MM8-DD4		Dusky			Kaikoura
MM9-HD1	8730	Hector's	Female	3	Pegasus Bay
MM8-HD1	9363	Hector's	Female	1	North of Christchurch
MM9-HD2	8736	Hectors'	Female	10	Pegasus Bay
MM9-HD3	8737	Hector's	Female	19	Pegasus Bay
MM9-HD4	8739	Hector's	Male	18	South of Timaru
MM10-HD1	8844	Hector's	Female	2	Pegasus Bay
MM10-HD2	8845	Hector's	Male	1	Akaroa
MM10-HD3	8846	Hector's	Male	1	North of Timaru
MM11-HD1	W004	Hector's	Male	1	North of Westport
MM11-HD2	W005	Hector's	Male	1	Westport
MM11-HD3	W007	Hector's	Male	1	Granity

Appendix 2. Concentrations of individual PCB cogeners (ng/g) in New Zealand marine mammals.

Congener	MM8-HD2	MM8-DD1	MM8-DD2	MM8-DD4	MM9-HD1	MM8-HD1	MM9-HD2	MM9-HD3	MM9-HD4	MM10-HD1	MM10-HD2	MM10-HD3	MM11-HD1	MM11-HD2	MM11-HD3
PCB#77[a]	66	52	14	3.3	47	64	35	61	190	30	40	70	110	170	70
PCB#126[a]	1091	53	65	5.1	1032	667	121	1000	740	400	600	800	130	150	250
PCB#169[a]	393	37	109	14	95	95	39	170	140	100	110	160	50	70	90
PCB#28	5.46	2.58	2.6	0.96	27.3	12.1	3.11	15.2	9.2	9.09	7.43	9.93	6.4	9.1	3.98
PCB#52	13.0	34.0	20.4	5.38	19.8	13.0	1.41	15.6	16.6	8.41	11.6	17.1	4.34	4.64	3.39
PCB#101	58.0	83.2	43.1	6.93	58.3	30.2	4.92	41.6	30.6	27.1	25.1	34.7	7.07	8.12	7.54
PCB#99	89.9	93.1	26.1	9.13	43.3	42.3	3.86	43.8	88.3	16.7	25.9	92.1	14.6	5.99	12.1
PCB#118	439	237	108	19.4	195	182	18.1	216	419	98.0	142	304	82.7	31.8	62.0
PCB#105	122	49.1	31.8	6.39	51.8	48.0	5.5	57.1	127	25.9	51.9	119	23.5	8.91	19.4
PCB#153	1495	1854	567	161	721	666	121	892	1587	300	199	n.d.[b]	170	87.1	133
PCB#138	1204	1154	427	124	519	458	86.2	592	1139	227	201	632	163	74.1	128
PCB#187	285	424	108	34.6	109	93.9	31.6	129	270	47.5	39.9	290	37.5	14.2	37.6
PCB#183	103	84.7	36.3	12.1	42.3	34.5	11.4	52.8	105	17.1	13.7	118	11.9	5.29	11.5
PCB#180	435	430	165	54.0	154	125	45.5	200	390	71.0	61.8	385	55.5	23.2	54.5
PCB#170	157	164	60.7	18.2	71.4	61.7	20.2	94.4	158	31	26.6	181	19.7	7.59	15.7
PCB#202	16.9	24.7	5.48	2.7	6.68	5.08	2.34	8.26	14.2	3.21	1.73	23.1	2.36	0.98	2.36
PCB#194	68.3	71.9	22.0	8.1	19.8	18.4	9.53	33.7	58.1	10.1	5.45	56.3	5.06	2.39	3.52
SUM (µg/g)	4.49	4.71	1.62	0.46	2.04	1.79	0.37	2.39	4.41	0.89	0.81	2.26	0.60	0.28	0.49
TE (pg/g)	175	40.2	23.9	3.94	131	92.5	15.5	120	136	54.4	81.4	130	24.9	20.2	34.8
Potency (µg TE/g PCB)	39	8.5	15	9	64	52	42	50	31	61	100	58	42	72	71

[a] Concentrations for PCB#77 PCB#126 and PCB#169 in pg/g. Concentrations for all other congeners in ng/g.
[b] n.d. = Not Determined due to mass spectrometer technical difficulties.

Appendix 3. Concentrations (pg/g) of individual PCDD and PCDF congeners in New Zealand marine mammals.

Congener	MM8-HD2	MM8-DD1	MM8-DD2	MM8-DD4	MM9-HD1	MM8-HD1	MM9-HD2	MM9-HD3	MM9-HD4	MM10-HD1	MM10-HD3
2,3,7,8-TeF	2.1	< 0.1	0.35	< 0.4	1.6	2.02	1.8	0.7	1.59	2.08	1.86
non-2,3,7,8-TeF	5.66	1.44	0.35	< 0.3	< 0.8	3.36	< 0.3	< 0.8	< 0.6	< 0.4	< 0.4
2,3,7,8-TeD	4.8	< 0.6	< 0.3	< 0.2	7.02	3.18	< 0.6	5.23	3.98	3.51	3.85
non-2,3,7,8-TeD	< 0.2	< 0.3	< 0.2	< 0.4	< 2	< 0.2	< 0.7	< 1	< 2	< 1	< 1
1,2,3,7,8-PeF	0.67	< 0.3	< 0.2	< 0.1	< 1	< 0.2	< 0.5	< 0.9	< 0.8	< 0.6	< 0.5
2,3,4,7,8-PeF	28.1	< 0.2	< 0.2	< 0.1	12.6	5.84	2.2	6.7	18.9	5.81	7.17
non-2,3,7,8-PeF	7.5	2.5	0.67	< 0.2	1.43	0.92	< 0.7	2.55	8.32	< 0.6	4.86
1,2,3,7,8-PeD	11.4	< 0.2	< 0.3	< 0.2	8.47	3.91	0.94	4.92	5.73	4.72	4.01
non-2,3,7,8-PeD	< 0.1	< 0.3	< 0.1	< 0.2	< 0.8	< 0.1	< 0.6	< 0.5	< 0.5	< 0.4	< 0.3
1,2,3,4,7,8-HxF	< 0.2	< 0.3	< 0.2	< 0.2	< 0.5	< 0.2	< 0.4	< 1	< 0.8	< 0.5	< 0.4
1,2,3,6,7,8-HxF	0.29	< 0.3	< 0.2	< 0.2	< 0.5	< 0.3	< 0.4	< 1	0.81	< 0.6	< 0.5
2,3,4,6,7,8-HxF	0.57	< 0.4	< 0.4	< 0.3	< 0.3	0.28	< 0.3	< 0.9	< 0.9	< 0.5	< 0.6
1,2,3,7,8,9-HxF	< 0.2	< 0.2	< 0.1	< 0.2	< 0.7	< 0.2	< 0.6	< 0.9	< 1	< 0.7	< 0.3
non-2,3,7,8-HxF	4.2	3.38	2.89	< 0.2	< 1	1.17	1.21	4.17	6.77	< 0.9	2.46
1,2,3,4,7,8-HxD	0.80	< 0.3	< 0.2	< 0.2	< 1	< 0.4	< 0.5	< 0.9	< 0.9	< 0.7	< 0.6
1,2,3,6,7,8-HxD	1.98	< 0.3	< 0.5	< 0.2	3.22	1.12	0.77	2.32	< 1	< 2.0	< 0.7
1,2,3,7,8,9-HxD	< 0.2	< 0.2	< 0.1	< 0.2	< 1	< 0.4	< 0.6	< 2	< 1	< 0.8	< 0.7
non-2,3,7,8-HxD	< 0.6	< 0.5	< 0.4	< 0.4	< 3	< 0.5	< 1	2.33	2.73	< 0.7	< 1
1,2,3,4,6,7,8-HpF	0.23	0.50	0.43	< 0.1	1.66	< 0.2	0.63	1.79	1.5	< 0.8	< 1
1,2,3,4,7,8,9-HpF	< 0.2	< 0.3	< 0.3	< 0.2	< 0.8	< 0.1	< 0.4	< 0.6	< 0.9	< 0.4	< 0.6
non-2,3,7,8-HpF	0.5	1.78	1.18	< 0.3	5.03	0.43	0.46	1.22	1.15	< 0.8	< 1
1,2,3,4,6,7,8-HpD	0.81	1.21	0.44	0.68	23.6	0.82	4.62	10.41	12.6	9.61	4.88
non-2,3,7,8-HpD	< 0.5	0.85	0.31	0.44	9.11	0.37	2.28	5.19	6.97	5.91	2.96
OCDF	< 0.7	1.3	< 0.7	< 0.6	15.3	< 0.6	< 2	< 3	< 3	< 4	< 3
OCDD	5.55	8.85	3.28	5.6	89.1	5.53	15.64	50.0	89.4	74.7	22.6
TE (pg/g)	25.2	0.5	0.4	0.3	18.7	8.50	2.32	11.9	17.1	9.42	9.91
Total TE (PCDDF+PCBs)	200	40.7	24.3	4.24	149.8	101	17.8	131.9	152.7	63.8	140
% TE from PCBs	87.4	98.7	98.2	92.9	87.5	91.6	87.0	91.0	88.8	85.2	92.9

Appendix 4. Comparison of total PCB concentrations in marine mammals from around the world.

Species	Reference	Location	PCBs (μg/g)
Bottlenose Dolphin	15	South Africa	13.8
Dall's Porpoise	8	North Pacific	8.6
White-sided Dolphin	8	Japan	37.6
Bottlenose Dolphin	16	East USA	81.4
Common Dolphin	16	East USA	36.5
White-sided Dolphin	16	East USA	50.1
Harbour Porpoise	17	U.K.	55.5
Dusky Dolphin	8	South of N.Z.	1.4
Baleen Whales[a]	3	N.Z.	< 0.05[c]
Minke Whales	18	West USA	3.3
Beaked Whales[b]	3	N.Z.	0.1 - 0.5[c]
Baird's Beaked Whales	19	Japan	3.0
Common Dolphin	3	N.Z.	0.75 - >1.0[c]
Hector's Dolphin	9	N.Z.	3.6

[a] Predominantly Minke Whale
[b] Predominantly Gray's Beaked Whale
[c] Measured as the sum of 16 predominant congeners.